



PATENT 1422-0509P

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

In Re Application of :
Masato FUJIKAKE et al. : Group Art Unit: 1713
Serial No.: 10/009,687 :
Filed: December 13, 2001 : Examiner: M. Reddick
For: CARBOXYLATED POLYMERIC COMPOSITION

DECLARATION UNDER 37 C.F.R. 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Masatoyo YOSHINAKA, residing at Himeji-shi, Hyogo-ken, Japan, hereby declares and states as follows:

1. That I am thoroughly familiar with the contents of said Application, its prosecution before the United States Patent and Trademark Office and the references cited therein.
2. That I am a graduate of Kansai University, Department of Engineering in the year 1994, majoring in Application department of chemistry.
3. That I have been employed in Sumitomo Seika Co., Ltd. in the year 1994 and have been assigned to the Functional Polymers Research Laboratory.
4. That I have been involved in the research and development of functional polymers.

5. That the following experiments were conducted by myself or under my direct supervision and control in order to prove that the carboxylated polymer composition of the present invention exhibits excellent physical properties unexpected from the process disclosed in USP 4,973,632(Nagasuna et al.).

EXPERIMENTAL:

Experiment I-1 (USP 4,973,632)

The same procedures of Example 1 as disclosed in USP 4,973,632 were repeated. The detailed procedures are as follows:

In a four-necked separable 2 L flask equipped with a stirrer, a reflux condenser, a thermometer, an inlet tube for nitrogen gas, and a dropping funnel was placed 1,000 ml of cyclohexane and dissolved 4.0 g of a sucrose fatty acid ester (DK-ESTER F-50, HLB=6, a product from DAIICHI KOGYO SEIYAKU Co., LTD.) and nitrogen gas was introduced into this solution to remove oxygen dissolved. In another flask containing a solution of 84.6 g of sodium acrylate, 21.6 g of acrylic acid, and 0.016 g of N,N'-methylene-bisacrylamide in 197 g of ion-exchanged water was dissolved 0.53 g of hydroxyethylcellulose (HEC-DAISERU EP-850, a product from DAISERU CHEMICAL Co., LTD.) and was prepared a monomer solution adjusted at a monomer concentration of 35 weight % and viscosity of 40 cps. To this monomer solution was dissolved 0.15g of potassium persulfate and then, nitrogen gas was introduced to remove oxygen dissolved in this aqueous solution.

Next, to the above separable flask solution was added the aqueous monomer solution in the latter flask and the mixture obtained was dispersed with stirring at 500 rpm. Then, polymerization reaction was initiated by raising bath temperature to 60 °C. and completed by maintaining this temperature for 2 hours. After polymerization, the reaction mixture was treated by an azeotropic distillation with cyclohexane to remove

water in the water-containing gel, and dried at 80 °C. under reduced pressure to obtain a polymer composition.

Experiment I-2

The same procedures as in Experiment I-1 were carried out except that 0.015 g of pentaerythritol tetraallyl ether was used in place of N,N'-methylene-bisacrylamide.

Experiment I-3

The same procedures as in Experiment I-1 were carried out except that 0.015 g of polyallyl saccharose was used in place of N,N'-methylene-bisacrylamide.

Experiment II-1

The same procedures as in Experiment I-1 were carried out except that 4.0 g of polyoxyethylene castor oil was used in place of the sucrose fatty acid ester.

Experiment II-2

The same procedures as in Experiment I-2 were carried out except that 4.0 g of polyoxyethylene castor oil was used in place of the sucrose fatty acid ester.

Experiment III (Present Invention)

The same procedures of Example 1 as disclosed at page 16, line 23 to page 17, line 15 of the present specification were repeated. The detailed procedures are as follows:

A 500-mL four-necked flask equipped with a stirrer, a thermometer, a nitrogen gas inlet tube and a reflux condenser was charged with 60 g of acrylic acid as an α,β -unsaturated carboxylic acid (a), 0.42 g of pentaerythritol tetraallyl ether as a compound

(b) having at least two ethylenically unsaturated groups, 3.0 g of polyoxyethylene castor oil (manufactured by NIKKO CHEMICALS CO., LTD. under the trade name of CO-3; adduct with 3 moles of ethylene oxide) as an alkylene oxide adduct of an ester (d) obtained from a polyhydric alcohol and a fatty acid, 0.009 g of azobisisobutyronitrile and 375 g of ethylene dichloride. The mixture was mixed with stirring, and thereafter nitrogen gas was blown into the solution at a flow rate of 100 ml/minute for 1 hour in order to remove oxygen gas existing in the upper space of the flask and oxygen dissolved in the resulting solution. Subsequently, a polymerization reaction was carried out for 3 hours by heating the solution to 70° to 75°C under nitrogen gas atmosphere.

After the termination of the polymerization reaction, the slurry formed was heated to about 110°C, and ethylene dichloride was distilled, to give 60 g of a carboxyl group-containing polymer composition in the form of white fine powders.

RESULTS:

As the physical properties of the products obtained in each Experiment, (1) dissolving time without stirring, (2) dissolving time with stirring, (3) viscosity of solution and (4) viscosity of neutralized viscous solution were evaluated in accordance with the methods described at page 17, line 21 to page 18, line 22 of the present specification. The results are shown in the following Table I.

Table I

Experiment No.	Dispersing agent	Crosslinking agent (g)
I-1	Sucrose fatty acid ester	N,N'-methylene-bisacrylamide (0.016)
I-2	"	Pentaerythritol tetraallyl ether (0.015)
I-3	"	Polyallylsaccharose (0.015)
II-1	Polyoxyethylene castor oil	N,N'-methylene-bisacrylamide (0.015)
II-2	"	Pentaerythritol tetraallyl ether (0.015)
III	Polyoxyethylene castor oil	Pentaerythritol tetraallyl ether (0.42)

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Experiment No.	Property	Dissolving Time		Viscosity of Solution (mPa•s)	Viscosity of Neutralized Viscous Solution (mPa•s)	Note
		Without Stirring (min.)	With Stirring (min.)			
I-1	Water-absorbent	—	—	—	—	USP 4,973,632
I-2	*	>180	>120	2000	2000	
I-3	*	>180	>120	2100	2100	
II-1	Water-absorbent	—	—	—	—	
II-2	*	>180	>120	2050	2050	
III	Soluble in water	20	3	2	59000	Present Invention

(Note)

* : Partly insoluble in water

DISCUSSION:

(1) According to the method disclosed in USP 4,973,632, as is clear from the results of Experiment No. I-1 shown in Table I, when sucrose fatty acid is used as a dispersing agent, and N,N'-methylene-bisacrylamide is used as a crosslinking agent, a water-soluble polymer cannot be obtained.

Therefore, even though the method disclosed in USP 4,973,632 is employed, a water-soluble polymer cannot be obtained.

(2) When pentaerythritol tetraallyl ether or polyallylsaccharose is used as a crosslinking agent in place of N,N'-methylene-bisacrylamide used in USP 4,973,632, as is clear from the results of Experiment Nos. I-2 and I-3, a partly water-insoluble polymer is obtained. The polymer is also inferior in water-solubility to the polymer obtained in Experiment III, and does not provide a neutralized solution having a high viscosity.

(3) When polyoxyethylene castor oil is used in place of sucrose fatty acid used in Experiment No. I-1, as is clear from the results of Experiment II-1 shown in Table I, a water-soluble polymer cannot be obtained.

Therefore, even though polyoxyethylene castor oil is used in place of sucrose fatty acid employed in USP 4,973,632, a water-soluble polymer cannot be obtained.

(4) When polyoxyethylene castor oil is used in place of sucrose fatty acid used in Experiment No. I-2, as is clear from the results of Experiment II-2 shown in Table I, a partly water-insoluble polymer is obtained. The polymer is inferior in water-solubility

to the polymer obtained in Experiment III, and does not provide a neutralized solution having a high viscosity.

(5) On the contrary, according to Experiment No. III, as is clear from the results shown in Table I, a water-soluble polymer which can be dissolved in water in a short period time and provides a neutralized solution having a high viscosity is obtained.

(6) The reason why a water-soluble polymer which can be dissolved in water in a short period of time and provides a neutralized solution having a high viscosity is obtained is thought to be based upon the following reasons.

According to Experiment No. III, since a specific crosslinking agent is used, and the polymerization is carried out in a non-aqueous system using for instance an inert solvent such as ethylene dichloride, a water-soluble polymer having the above-mentioned properties can be obtained.

On the contrary, according to Experiment Nos. I-1 and II-1, since N,N'-methylene-bisacrylamide disclosed in USP 4,973,632 is used as a crosslinking agent, and a specific crosslinking agent such as pentaerythritol tetraallyl ether is not used, a water-soluble polymer cannot be obtained.

Moreover, even though N,N'-methylene-bisacrylamide is changed to pentaerythritol or polyallylsaccharose as a crosslinking agent, as is clear from the results of Experiment Nos. I-2, I-3 and II-2, a water-soluble polymer having a high water-solubility and providing a neutralized solution having a high viscosity cannot be obtained, since the polymerization is carried out by a reverse-phase suspension polymerization process using water.

Accordingly, since Experiment No. III employs a specific crosslinking agent, and the polymerization is carried out in a non-aqueous system using for instance an

inert solvent such as ethylene dichloride, a water-soluble polymer having the above-mentioned properties can be obtained.

However, USP 4,973,632 does not disclose or suggest that the polymerization is carried out using a specific crosslinking agent in a non-aqueous system.

Therefore, there is no motivation in USP 4,973,632 to arrive at the present invention.

6. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

7. Further declarant saith not.

Masatoyo Yoshinaka

Masatoyo YOSHINAKA

September 13, 2004

Date